Chemical Schemes for Surface Modification of Icy Satellites: A RoadMap

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Abstract

The icy Galilean satellites of Jupiter are subject to magnetospheric plasma ion bombardment which induces chemical changes within the ice. The possible detection of CO_2 on the surface of Ganymede by the Galileo spacecraft makes for a more complicated chemistry and increases the number of chemical compounds that may then be present. We outline chemical schemes for the irradiation of pure and mixed ices H_2O/CO_2, and suggest species which observers may detect on Europa, Ganymede and Callisto, such as C_3O_2, H_2CO_3, H202, C03, H02, CO, H2C0, CH2C0, KOH, K20 and others. Column abundances of compounds in the ice are calculated using a specified energy input and G values for estimating yields.

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The icy satellites of Jupiter have been generally understood to consist of mostly water ice on their surfaces. Observations by ground-based observers (e.g., Calvin et al 1995) show an almost pure water ice spectrum. Other materials thought to be present are oxygen molecule (O_2) on Ganymede (Spencer, et al 1995), SO_2 on Europa (Lane et al 1981, Nell et al 1995) and ozone (03) on Ganymede (Nell et al 1996), as well as non-volatiles such as clays, and sulfur compounds.

The Galileo spacecraft has recently inferred the possible presence of other species on the surfaces of the icy satellites. A band at 4.25 micron was seen in the spectrum of Ganymede taken by the NIMS instrument (Carlson 1996). This band also occurs in the spectrum of Callisto (Carlson et al. 1996b), and is a weak band in Europa's spectrum (Carlson 1996). This band could result from the presence of $C0_2$ or of hydroxylated minerals on their surfaces. This detection adds a new dimension to the mix of chemical species on the surfaces, and leads to speculation of other molecules that can then be predicted to occur there. The surfaces are subject to continuous magnetospheric bombardment from plasma sources which will induce chemical reactions in the solid state. If CO_2 is present, a whole plethora of species could then be present, and should be sought by observers.

This paper is therefore attempting to provide a roadmap for observers of the chemistry that could result from a water ice surface containing CO_2 that is bombarded with magnetospheric plasma ions over the age of the solar system.

PLASMA SOURCES

Jupiters magnetosphere has been examined in depth by Pioneer, Voyager, Galileo and Ulysses spacecraft. The plasma populations known to represent include the following species:

H^+, He^+, H_2^+, H_3^+, O^+, O^{2+}, 03+, O^{4+}, O^{6+}, S^+, S^{2+}, S^{3+}, S^{4+}, S^{5+}, S_2^+, SO_2^+, Na^+, K^{2+}, C^{6+}, H_2O^+, H_3O^+, OH^+

The oxygen and sulfur species are thought to come from volcanic emissions on Io, which deposit many tons of material per minute into the magnetosphere (Bagenal, 1991, Geiss et al 1992). Sodium and potassium species are also from Io volcanic emissions. Water group ions (H_2O^+, H_3O^+) come from sputtering of the water surfaces on Europa, Ganymede and Callisto (Schreier, et al 1993). The solar wind supplies species such as H^+, He^+, O^{6+} and C^{6+} . Jupiter's ionosphere may be the source of H^+, H_2^+, and H_3^+ (Geiss et al 1992). Therefore, we can use as impactors all of these species onto the surfaces of the satellites. Some will implant and cause chemical reactions with themselves (e.g. Lane et al 1981). Other plasma impactors will simply act as battering rams, and reshuffle the atoms in the molecules that are present, thereby inducing new chemistry.

SOLID STATE CHEMISTRY

Figure 1 shows pathways for chemistry in water irradiated in the solid state. Many lab experiments have been performed to study this system and we summarize the products here. When water is bombarded with ions or electrons, or UV radiation, a first result is to create H and OH. OH may then be dissociated into O and H. These fragments are constrained inside a void in the lattice, and so will react quickly together to form H_2 and O_2 (Benit et al 1990). O will require a 3rd body, (such as another O, a neighboring H_2O or the wall of the lattice void) to react with another O to make O_2. When combined, the 0_2 formed will be in an excited state (Matich et al 1993), and will radiate or transfer its energy to the wall of the void. H_2 will diffuse out of the lattice, leaving 0_2 as a fractionating product.

The diffusion of O atoms through the lattice to form O_2 was demonstrated by Benit et al (1990) who overlaid 2 different isotopic forms of water, $H_2{}^{16}O$ and $D_2{}^{18}O$ and bombarded the two layers with Ne^+ ions. They found all three possible combinations of 0_2, that is, $^{16}O_2$, $^{18}O^{16}O$, and $^{18}O_2$, showing that O travelled around the lattice from one layer to another, and was mixed, with the impacting ion acting as a ram to reshuffle the atoms. O_2 has already been observed on Ganymede (Calvin et al 1996). O_2 will combine with another constrained O and can form O_3. (See Figure 1) Ozone has been detected on Ganymede's surface as well (Nell et al 1996).

Ozone is easily dissociated by UV light and ion radiation, yielding excited state oxygen atoms, $O(^1D)$. If radiation is hard enough, possibly a higher excited state, $O(^1S)$, might form. The lifetime of $O(^1D)$ against radiating to the ground state is long (-100 see); therefore it will likely react with a nearby H_20 molecule first, before it has time to emit a red photon (emission at 6300 A). The lifetime of 0(1 S), however, is short (-1 see) and so it may emit a green photon (at 5577 A) to decay to $O(^1D)$, or a UV photon (at 2972 A) before it has time to do much chemistry. However, $O(^1S)$ can also react with water, yielding two OH molecules (Zipf 1969). If it radiates at 2972 A, the O atom will then be in the ground state, $0(^3P)$, and can still combine with another O to make 0_2.

A product of $O(^1D)$ reaction with water will be hydrogen peroxide (HOOH). (Sedlacek and Wight, 1989). Another reaction forming HOOH (or H_2O_2) is the combination of 2 OH radicals, which will be present from the initial water fragmentation. Neftel et al (1984) showed that HOOH is easily made from cosmic ray fluence into water ice at the South Pole on Earth. They estimated a cosmic ray flux attenuation of 0.023 per meter in the first 100 meters of ice in the South Pole ice sheet, and assumed an ion-pair production rate of $5x10^3$ pairs $g^{-1}s^{-1}$ in the ice. They found that at least 5 ppb of H_2O_2 could have been formed in the ice over a 1500 year period. They also determined that H_2O_2, though quite reactive, has a long lifetime in polar ice samples. It should also occur upon bombardment of the much colder ices of Europa, Ganymede and Callisto, and be retained longer.

Johnson and Quickenden (1997) discuss radiolysis of H_2O ice on the satellites. They give G values for production of stable products from irradiation from pure water. G values are a measure of the amount of a given species produced for each 100 eV of energy deposited into the ice. In Johnson (1990), the G values for production of products from pure CO, CO_2 and O_2 are given. The column density of a chemical species formed can be calculated from a simple equation relating energy input (E) , yield (G value) and time (Delitsky and Thompson 1987). This equation is:

$$N = EGt$$

The energy input rate is calculated from Mauk et al (1996). They give energy intensity of hot ions as a function of radial distance from Jupiter. For Europa, at a distance of 9.6 Rj, the energy input is 2.9×10^{10} keV/cm^{2}sec. For Ganymede, the input would be 1.5×10^{10} keV /cm2sec. Mauk et al. do not plot the energy out to Callisto's distance, but extrapolating their Figure 3, we can estimate an energy input rate as being - 5×10^{9} keV/cm2sec. Using these G values and energy flux, we have calculated the quantities of radiation products from the pure materials, H_2O and CO_2. We have also estimated amounts of products that form from irradiation of mixed ices H_2O/CO_2.

Table 1 gives the G values, and our calculation of column density, N, that will be formed over -1000 years from this plasma bombardment. As an example, for the formation of H_2O_2, we have a G value of 0.4 molecules/100 eV. Using the energy input rate given, the column density of H_2O_2 formed is then 3.65×10^{21} molecules/cm^2. This is equal to 0.206 g/cm^2.

Column densities for Europa are calculated in Table 1. For Ganymede and Callisto, a multiplicative factor can be used to calculate abundances which is a ratio of the energy input rates given above.

H_2O_2 (or HOOH) will react with any OH available in the lattice void to form the radical HO_2. This molecule was found in irradiated ice experiments by de Vries et al. (1984). HO_2 will react with itself to give back HOOH, which may then react with OH to reform the HO_2. HO_2 has a peak absorption in the UV near 210 nm. From this chemistry, it is possible that HO_2 may also be detected on the surfaces of the icy Galilean satellites.

Figure 1 then summarizes the hydrogen-oxygen species that are products of the irradiation of H_20 ice. The addition of other species to the mix, such as the CO_2 suggested to be present on Ganymede by the NIMS experiment on Galileo (Carlson et al 1996) makes the chemistry even more interesting and diverse.

Figure 2 shows the various carbon-hydrogen-oxygen species that could be present on Ganymede, if a H_2O/CO_2 ice is subjected to plasma radiation from Jupiters magnetosphere. Benit et al (1988) used 2 different ratios of H_2O to $C0_2$ in their ice mixtures, 2:1 and 0.2:1 H_2O/CO_2. These were bombarded with He^+ ions, with the ices at 77 K. They found evidence in the IR for species such as CH_2C0 (ketene), H_2CO (formaldehyde), CO, C_3, OH, O_3, CH_4, HCO and C03.

Irradiation of H_2O/CO_2 mixtures was also done by DelloRusso et al (1993). They found evidence for H_2CO, and also H_2CO_3 (carbonic acid). Its structure is $(OH)_2CO$. They give a G value for formation of carbonic acid from proton bombardment of H_2O/CO_2 as G = 0.5 /100 eV. A G value for formation of H_2CO from the irradiation of H_2O/CO mixtures is given as G = 1.0 /100eV. We assume this value also applies to the yield of H_2CO from H_2O/CO_2. Table 2 shows amounts of products that form from irradiation of mixed ices H_2O/CO_2, estimating G values for them, if not otherwise available. H_2CO forms a complex with $C0_2$ at low temperature and its absorption in the IR will be strongly blue-shifted, relative to free H_2CO. (van der Zwet, et al. 1989)

The presence of CO is quite interesting and if it accumulates into pure crystals or occurs as a clathrate, plasma irradiation will eventually lead to carbon suboxide (C_3O_2), a linear molecule which has been proposed to be on the surface of Mars (Plummer and Carson, 1969, Oyama et al 1977) , and in the ices of Comet Halley (Huntress and Delitsky 1988, Huntress Allen and Delitsky 1991). A small amount of C_30_2 was in fact formed upon proton irradiation of H_2O/CO_2 (Moore et al (1991)). It will rapidly hydrate to malonic acid ($HOOC$-CH_2-$COOH$) in water solution (Smith et al 1963), or it will polymerize (to $(C_3O_2)_n$); its behavior in solid state water ice solution has not been determined. (Allen et al 1995).

Formation of CO usually occurs upon bombardment of CO_2, and formation of CO_2 occurs upon bombardment of CO. Therefore, there is a back and forth processing of CO <--> CO_2 and the detection of one implies the formation and existence of the other over time. Chemical products of H_2O / CO irradiation are also then worth investigating. Other species formed in H_2O/CO mixtures were HCO, C03, C_2O and CH30H ((Moore et al (1991)).

Many of these species should have a long lifetime in the ice. Temperatures on the surfaces of the Galilean satellites are between 80- 130 K. Many of these compounds are below their melting points in this region. For example, H_2CO has a melting point of 181 K. H_2O_2 melts at 272 K. 03 has a melting point of 80 K, and a boiling point at 162 K (at STP). Therefore, it may find itself in the liquid state if formed in a water ice lattice at 100 K. Ketene, an unstable radical, may eventually react with water to yield acetic acid, CH3COOH, as a stable product (Allen et al. 1995), whose melting point is at 289 K.

Other channels of chemistry from ion implantation are shown in Figure 3. Sulfur ions of charge +1 to +5 exist in the Jovian magnetosphere and upon impact into the water ice matrix on an icy satellite should form SO and SO_2 (Lane et al 1981, Nell et al 1995). Lane et al noted the presence of an absorption near 280 nm which they attributed to SO bonds on Europa's surface. Nell et al (1995) thought the spectra were more characteristic of SO_2. If $S0_2$ is present, addition of O atoms will form SO_3.

Experiments by Sack et al (1992) showed that when S ions are impacted into water, a band in the range of 260-280 nm occurs in the reflectance spectrum. (However, a band in this region also occurred when they bombarded the water ice with other ions such as argon and helium, Ar^+ and He^+. This implies that the band is more a function of some product of water fragmentation (perhaps a solid state spectrum of OH in a broken ice lattice), rather than conclusive proof that it results from an SO bond. The 1-1 band at 2811 A in the OH gas phase spectrum could theoretically be broadened by being immersed in a solid state lattice and might provide contributions to this feature. Spectra of other water fragment products such as HO_2 and H_2O_2 show no features, or only a smooth change in this region (Okabe, 1978). The recorded spectrum in Sack et al. is of modest resolution and not sufficiently detailed to provide a feature shape which enables discrimination of sulfur products from water products. If sulfur is present, a definitive band arises from the production of S-O or SO_2, which are not distinguishable from each other with low resolution reflection spectroscopy. The most probable condition for the Galilean icy satellites is that both conditions co-exist, namely S-0 / $S0_2$ and fragmented H_20 and OH all comingled in a disturbed water ice lattice. Further work is needed to clarify this.)

Other ions bombarding the surfaces are sodium and potassium ions. Their presence was determined by the Voyager PLS instrument (McNutt 1993). McNutt found that potassium is doubly charged, K^{2+}. Sodium is as Na^+. K^{2+} was detected at 10 R_J (near Europa's distance) and out at 40 R_J, beyond the orbit of Callisto. Neutral sodium forms an extensive cloud in the Jovian system, which is spewed out from 10, and spirals out to great distances (Smyth 1992). Brown and Hill (1996) measured the D line of sodium emission near Europa out to 20 Europa radii. The neutral sodium from Io will be ionized rapidly, and some ions will act as impactors onto Europa's surface. Brown and Hill suggest that sodium salts such as Na_2O and NaOH are possible surface products. The neutral sodium emission near Europa may result from these products. From McNutt's suggestion of doubly charged potassium, probably other candidate materials formed in the ice would be K_2O and KOH.

Abundances of K and Na ions are given by McNutt (1993) as being 10-15 % that of the main ions O+ and S+, and comparable to the amounts of water group ions, such as H_2O^+. Therefore they could make a contribution to the chemistry. Abundances of KOH, K_2O, NaOH and Na_2O (as well as SO and SO_2) are calculated in Table 3 using energies and number densities from McNutt (1993). For example, a number density of 3.7 cm^{-3} was given for K^{2+} ions at Europa's distance (near 10 R_J) as found by Voyager 2. Using a coronation velocity of 100 km/see, this yields a deposition flux of 3. $0x10^7 cm^{-2}$ see-l. To build up a column density of $1x10^{16}$ ions/cm^2 as deposited in the first centimeter of surface would take - 10 years. Over a 1000 year period, one would get a column of $1x10^{18}$ ion/cm^2. For KOH, this is 0.000092 g/cm^2. Column abundances are calculated for Europa. See Table 3.

At Ganymede, the coronation velocity is 150 km/see, and the ion densities are $^1/_8$ what they are at Europa. Therefore, to get the abundances at Ganymede, values are multiplied in Table 3 by $^{150}/_{100} \times {^1/_8} = 0.187$. For Callisto, coronation velocity is -200 km/see and ion densities are on the order of - $^1/_{370}$. This is calculated from relative densities of K^{2+} given in McNutt (1993), as measured by Voyager at 10 R_J and 40 R_J. Therefore, for Callisto, values are multiplied by $^{200}/_{100} \times {^1/_{370}} = 0.0054$.

Figure 4 shows ions that were detected in an ion mass spectrometer when water ice was sputtered using Ar‘, He^+ and H_2^+ ions (Haring et al 1984). Products such as OH+, 02+, 0+, H_2O^+, HO_2^+ and $H_2O_2^+$ were seen, as well as cluster ions such as $H^+(H_2O)$, $H^+(H_2O)_2$, and $(H_2O)_2^+$. Clusters were also observed by others in sputtered beams. Benit et al (1986) noted clusters of $H(H_2O)_n^+$ where n maybe as high as 20 (i.e. 20 water molecules are attached to H^+ ion, the cluster being held together by van der Waals' forces).

Clusters were noted by de Vries et al(1984), which were sputtered along with neutral products 0_2, HO_2, OH, and O. Schreier et al (1993) modelled a Europa torus and many of these ions are assumed in their model to come from sputtering of water ice on Europa's surface.

SUMMARY

Therefore, assuming the 4.25 micron band detected on Europa, Ganymede and Callisto is indicative of the presence of CO_2, we predict the occurrence in the water ice surfaces of these satellites the following molecules :

CH_2CO, H_2O_2, HO_2, H_2CO, H_2CO_3, OH, K_2O, KOH, S03, C03, CO and C_3O_2.

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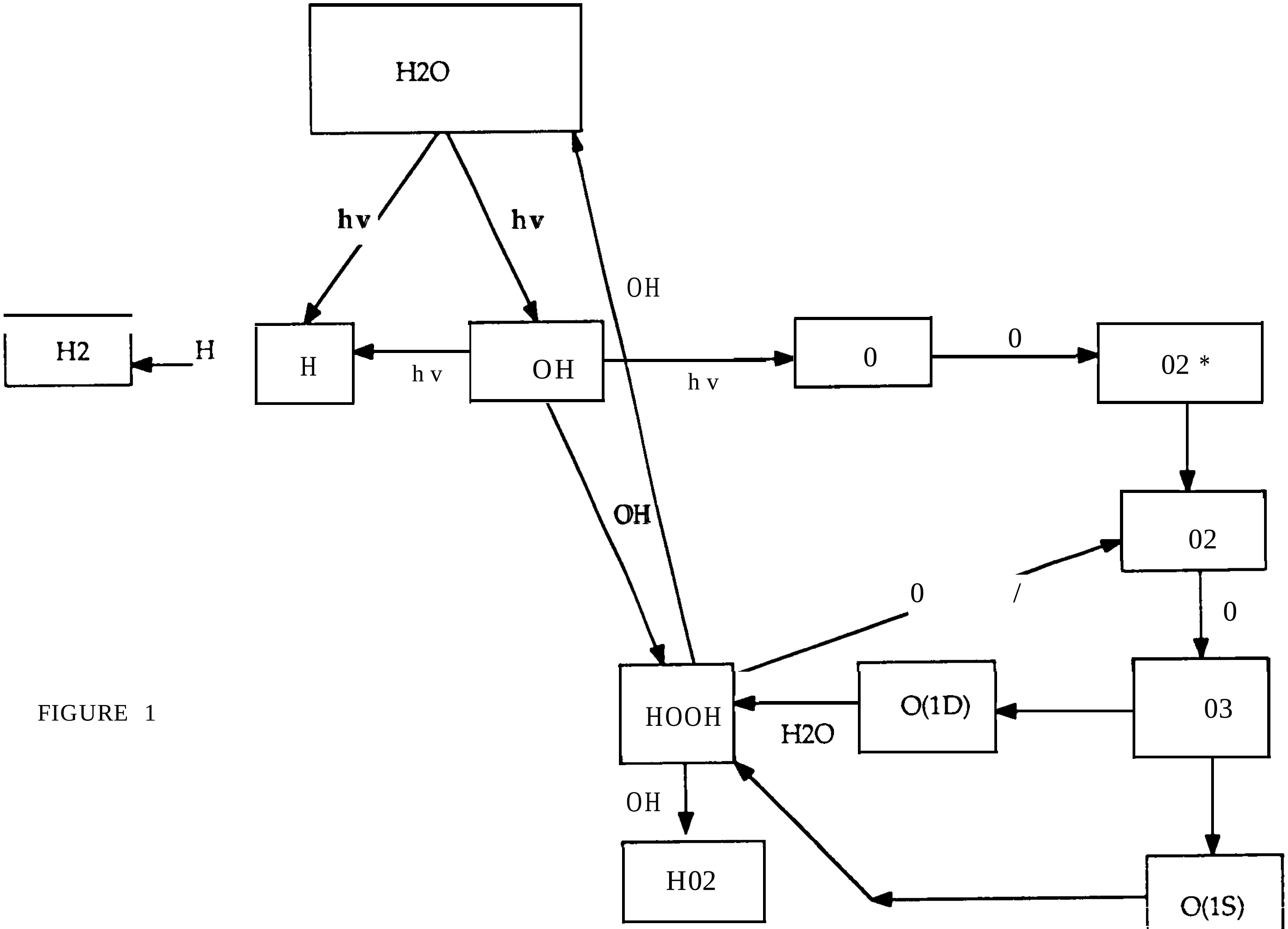
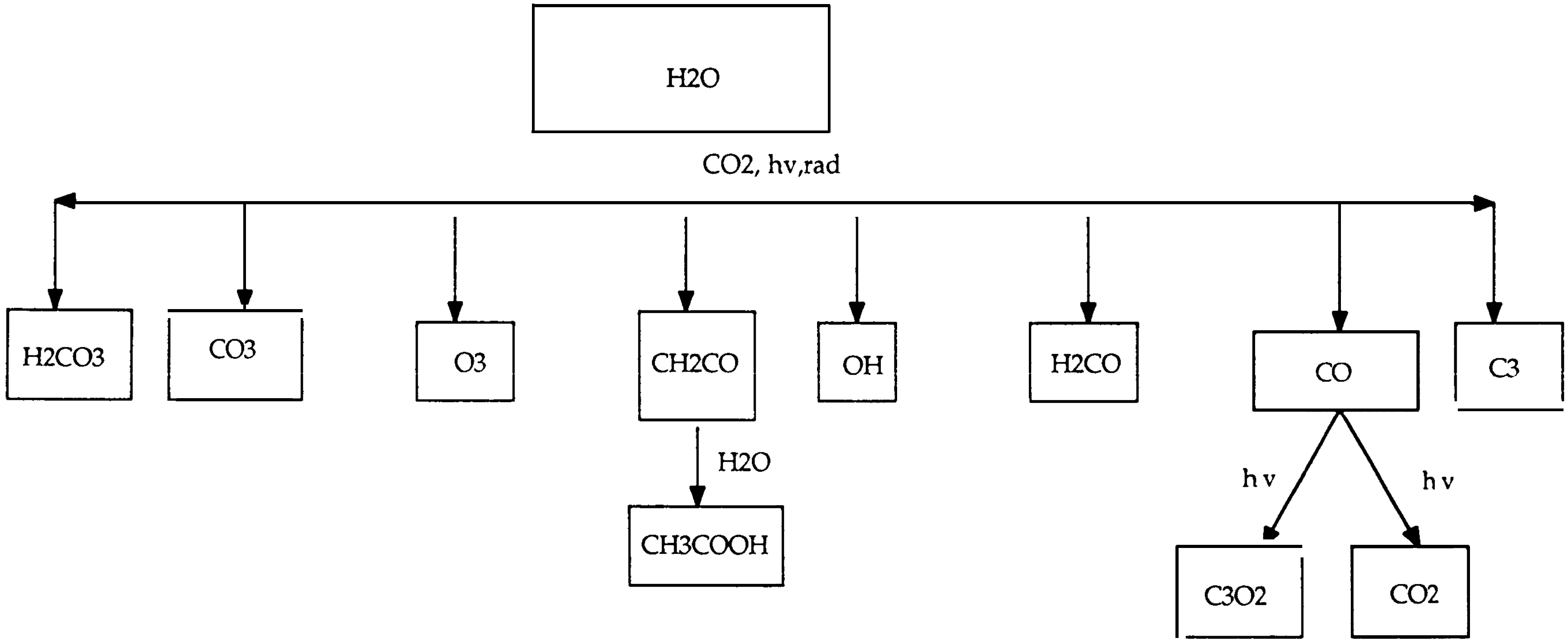


FIGURE 1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CO2, hv,rad
H2CO3
CO3
O3
CH2CO
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CH3COOH
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H2CO
CO
h v
h v
C3O2
CO2
C3

FIGURE 2

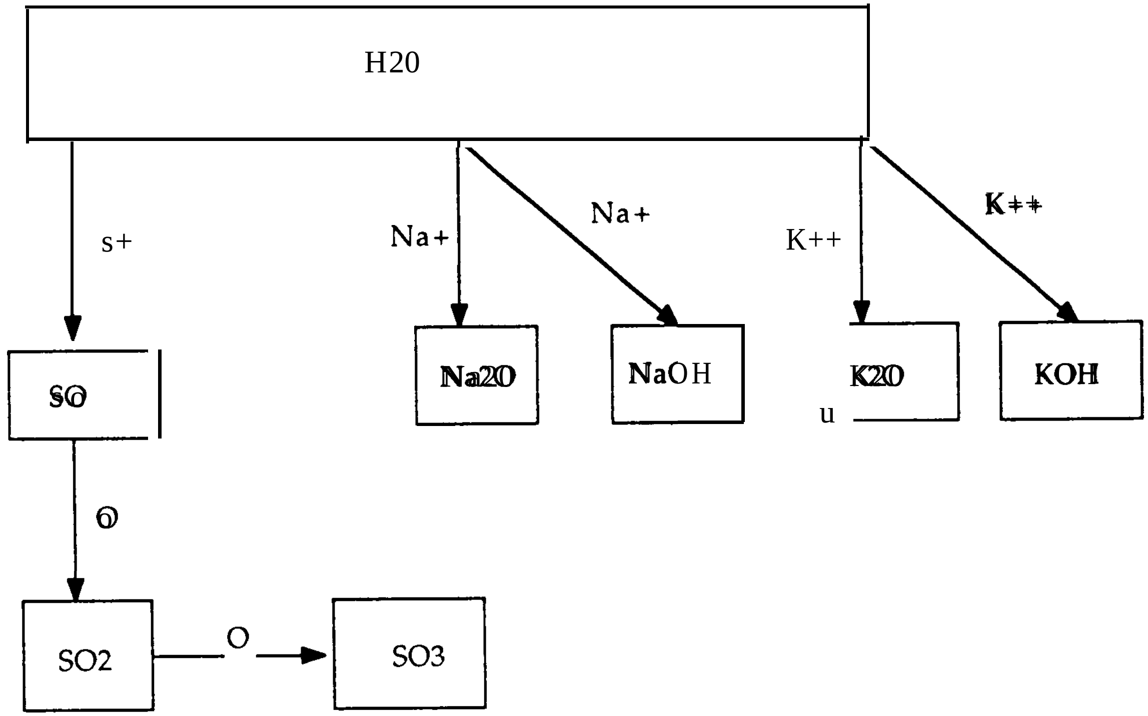

FIGURE 3

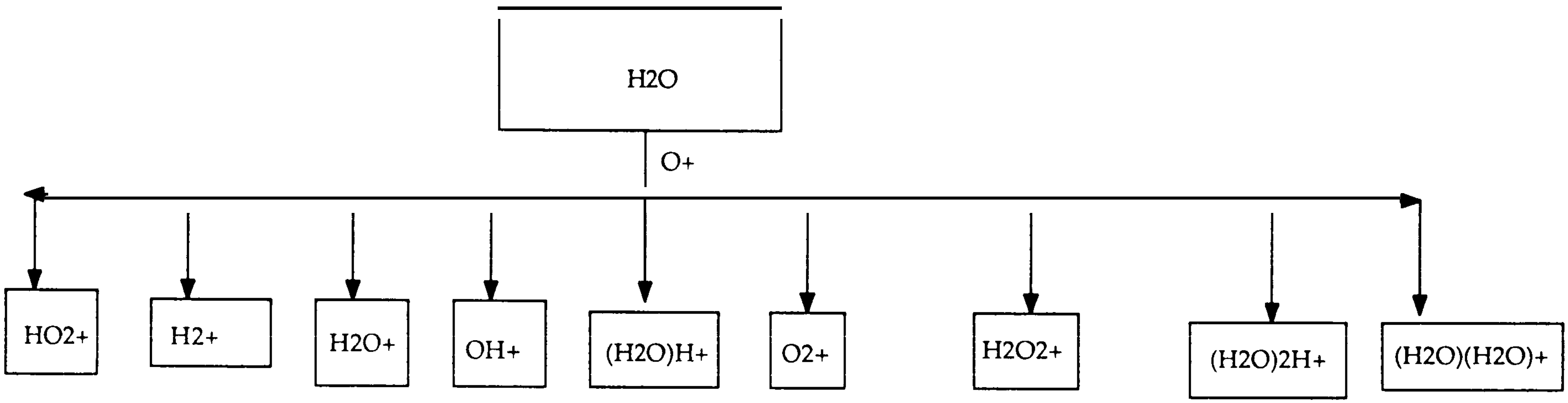

FIGURE 4

FIGURE CAPTIONS :

FIGURE 1
Chemical schemes for irradiation of pure water ice.
Some of these reactions may also occur from UV photolysis.

FIGURE 2
Chemical compounds that result from plasma ion bombardment of mixed ices H_2O/CO_2. Continued processing may eventually lead to polymerization.

FIGURE 3
Chemistry arising from implantation into water ice of common Jovian magnetosphere ions, such as S^{n+}, Na^+ and K^++

FIGURE 4
Ions formed by bombardment and sputtering of water ice by Jovian plasma ions. As derived from Haring et al. (1984)

TABLE 1

Products of Irradiation of Water Ice and CO_2 Ice (Pure Materials)
Yield over 1000 years

Species	G	N (molec/cm^2)	L (g/cm^2)
H_2O_2	0.4	3.65x1021	0.2064
HO_2	(0.4)	3.65x1021	0.2003
OH	0.3	$2.74x10^{21}$	0.0771
O_2	0.15	1.37X1021	0.0725
H_2	0.7	6.38x1021	0.0212
03 (from O2)	9.0	$8.22x10^{22}$	6.554
CO (from CO2)	9.0	$8.22x10^{22}$	3.822

TABLE 2

Products of Irradiation of H_2O / CO_2 Mixed Ices
Yield over 1000 years

Species	G	N (molec/cm^2)	L (g/cm^2)
H_2CO_3	0.50	$4.55x10^{21}$	0.4703
H_2CO	1.0	9.13X1021	0.4524
CH_2CO	(0.50)	$4.55x10^{21}$	0.3175
CO_3	(0.05)	4.55x102O	0.0452
HCO	(0.05)	4.55x102O	0.0217
C_3O_2	(0.50)	$4.55x10^{21}$	0.5141

Notes:

1) G values for pure materials are from Johnson (1990) and Johnson and Quickenden (1997); G values for mixed ices are from DelloRusso (1993); Ozone yield is from pure 0_2 (which is created from H_2O radiolysis). CO yield is from pure CO_2. Values in parentheses are assumed values; Ketene is assumed to be similar to H_2CO_3 in G value, C03 and HCO are assumed to be one tenth yield of H_2CO, based on relative intensities of IR peaks in experiments by Benit et al (1988).

2) This table gives values for Europa. To get abundances for Ganymede, multiply the values in the Table by 0.517. To get abundances for Callisto, multiply the values in the Table by 0.172. These fractions are the ratios of energy input, as derived from the paper by Mauk et al (1996). See text.

TABLE 3

Products of Ion Implantation of K^{2+}, Na^+ and S^+ into Water Ice
Yield over 1000 years

Species	I.D. (cm^{-3})	N ($molec/cm^2$)	L (g/cm^2)
K_2O	3.72	1.0×10^{18}	0.000092
KOH	3.72	1.0×10^{18}	0.000156
Na_2O	(3.72)	1.0×10^{18}	0.000102
NaOH	(3.72)	1.0×10^{18}	0.000066
SO_2	19.7	5.29×10^{18}	0.000560
SO	19.7	5.29×10^{18}	0.000421

Notes:

1) I.D is ion number density , from McNutt (1993), for K and Na ions measured at 10 R_J (near Europa's distance). For K-containing products, ion density refers to K^{2+} as the implanting ion. For Na-containing products, ion density refers to Na^+ as the implanting ion. Ion densities are not given in McNutt (1993) for Na^+ at 10 R_J; however, the K^{2+} and Na^+ densities are almost equal at 40 R_J , as given in McNutt (1993), so we assumed $Na^+ - K^{2+}$ at 10 R_J for the purposes of our calculation. For S-containing products, ion densities are for total S ions, as given in Bagenal (1994). A coronation velocity of 100 km/see is used to derive the flux of ions to the surface of Europa.

2) This table gives values for Europa. To get abundances for Ganymede, multiply the column densities in the Table by 0.187. To get abundances for Callisto, multiply the column densities in the Table by 0.0054. These satellites have higher coronation velocities, but lower ion densities. See text.